

INTERMEDIATE ENERGY NUCLEAR FISSION

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av

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Abstract Nuclear fission has been investigated with the double-kinetic-energy method using silicon surface barrier detectors. Fragment energy correlation measurements have been made for U, Th and Bi with bremsstrahlung of 600 MeV maximum energy. Distributions of kinetic energy as a function of fragment mass are presented. The results are compared with earlier photofission data and in the case of bismuth, with calculations based on the liquid drop model. The binary fission process in U, Yb, Tb, Ce, La, Sb, Ag and Y induced by 600 MeV protons has been investigated yielding fission cross sections, fragment kinetic energies, angular correlations and mass distributions. Fission-spallation competition calculations are used to deduce values of macroscopic fission barrier heights and nuclear level density parameter values at deformations corresponding to the saddle point shapes. We find macroscopic fission barriers lower than those predicted by macroscopic theories. The total kinetic energies at symmetric mass division are corrected for the effects of neutron evaporation after fission and found to be in good agreement with Viola's empirical relationship. No indication is found of the Businaro-Gallone limit expected to occur somewhere in the mass range $A = 100$ to $A = 140$. For Ce and La asymmetric mass distributions similar to those in the actinide region are found. A method is described for the analysis of angular correlations between complementary fission products. The description is mainly concerned with fission induced by medium-energy protons but is applicable also to other projectiles and energies. It is shown that the momentum and excitation energy distributions of cascade residuals leading to fission can be extracted.	
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INTERMEDIATE ENERGY NUCLEAR FISSION

by

Gunnar Hyltén

This thesis is based on the following papers:

- I. "Photofission in ^{238}U , ^{232}Th and ^{209}Bi at Intermediate Energy", Z. Physik A288 (1977) 333 (in collaboration with Mats Areskoug, Hans-Åke Gustafsson and Bent Schrøder)
- II. "Analysis of angular correlations between complementary fission products", Nucl. Instr. 163 (1979) 165 (in collaboration with Gert Andersson, Mats Areskoug, Hans-Åke Gustafsson, Bent Schrøder and Einar Hagebø)
- III. "Binary fission in Tb and Ag by 600 MeV protons", Phys. Lett. 64B (1976) 421 (in collaboration with Gert Andersson, Mats Areskoug, Hans-Åke Gustafsson, Bent Schrøder and Einar Hagebø)
- IV. "Mass yields in the binary fission of Ag, La and Tb", Phys. Lett. 71B (1977) 279 (in collaboration with Gert Andersson, Mats Areskoug, Hans-Åke Gustafsson, Bent Schrøder and Einar Hagebø)
- V. "Medium Energy Proton Induced Fission in Tb, La and Ag", Z. Physik A293 (1979) 241 (in collaboration with Gert Andersson, Mats Areskoug, Hans-Åke Gustafsson, Bent Schrøder and Einar Hagebø)

- VI. "Fission of light and medium-heavy nuclei induced by 600 MeV protons", Proceedings of the International Symposium on Physics and Chemistry of Fission, Jülich, 1979 (International Atomic Energy Agency, Vienna, Austria, 1980), Vol. II, p. 329 (in collaboration with Gert Andersson, Mats Areskoug, Hans-Ake Gustafsson, Bent Schrøder and Einar Hagebø)
- VII. "Mass distributions in the fission of medium-heavy and light nuclei", Phys. Rev. C24 (1981) 769 (in collaboration with Hans-Ake Gustafsson, Bent Schrøder and Einar Hagebø)

SUMMARY

Introduction

The exploration of the nucleus is a delicate task as it can only be made by indirect observations. So nuclear experiments depend strongly on the techniques being available. The general technical improvements have quickly been introduced to the field of nuclear science often in very specialized forms. The progress in accelerators, detectors, electronics and computers made the experiments presented here feasible.

Fission

The liquid drop model, according to which the nucleus is like an incompressible and uniformly charged liquid drop, was used by Bohr and Wheeler (Ref. 1) in their account of the mechanism of nuclear fission. If a nucleus (liquid drop) for some reason is disturbed from its equilibrium form, the Coulomb energy will decrease, while the surface energy will increase. For stable nuclei the change in Coulomb energy is smaller than the change in surface energy as a function of deformation. So we have a fission barrier. (See fig. 1.)

For increasing element number the Coulomb energy increases faster than the surface energy. So if we try to produce nuclei with more and more protons we will reach a point where the fission barrier vanishes. Such a nucleus will undergo immediate fission.

Classically we have either a fission barrier and no fission or no barrier and immediate fission. Quantum mechanically we can have penetration of the barrier by the tunnel effect and thus have spontaneous fission. The larger the barrier, the lower the probability of fission and the longer the half-life.

It is possible to increase the fission probability by giving the nucleus excitation energy. Thus bombarding nuclei with particles may induce fission. If the excitation energy is low, the nucleus may

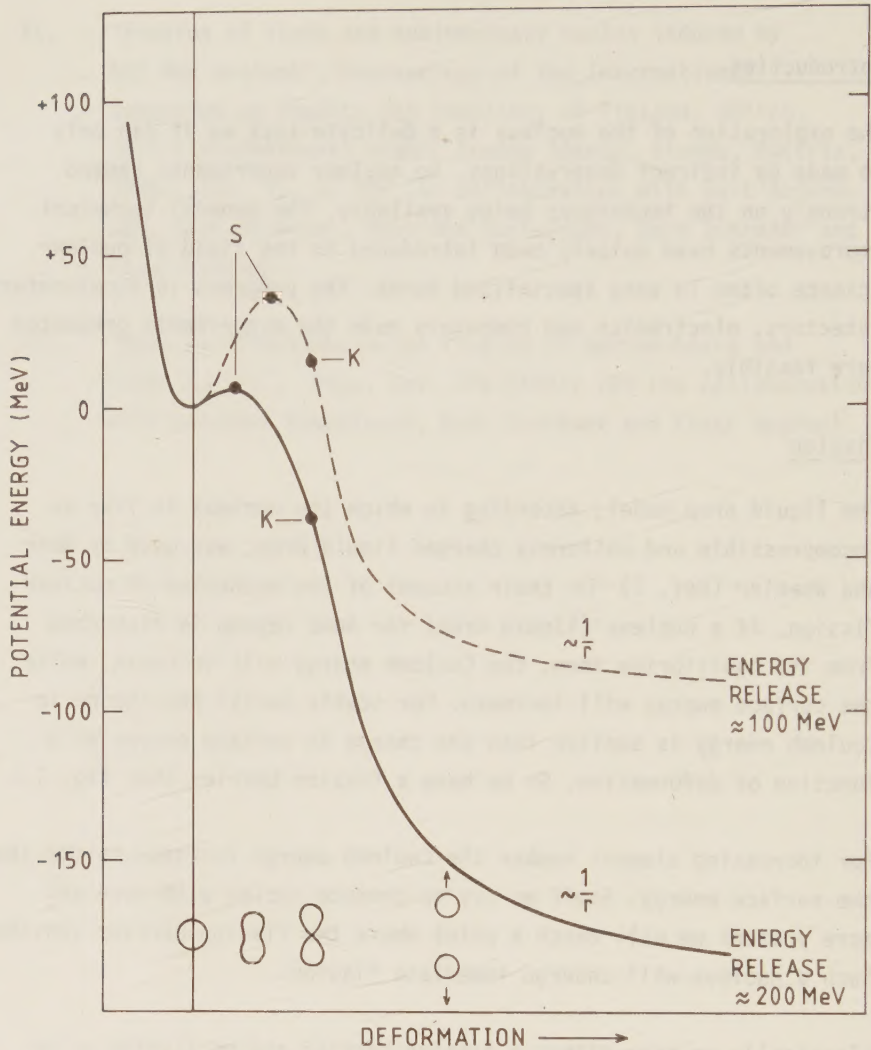


Figure 1.

Fission barriers according to the liquid drop model. The solid curve is for a nucleus in the uranium region ($A \sim 240$) and the dashed curve is for a nucleus in the rare earth region ($A \sim 165$). S indicates the maximum of the barrier (the saddle point). K indicates the scission point, i.e. the point where the nucleus is divided into two fragments. (From Wene, ref. 2).

still be below the fission barrier and decay by tunnelling. What increases the fission probability is a combination of increased level density and a thinner barrier to penetrate. For higher energies we come above the barrier. In both cases fission competes with other modes of deexcitation.

For heavy nuclei the liquid drop model predicts symmetric mass distributions. This is because it gives higher potential energy (and thus a higher barrier) for asymmetric than for symmetric deformations. But spontaneous fission and low energy induced fission of nuclei in the actinide region give experimental mass distributions which are highly asymmetric. (As an illustration fig. 2 shows examples of asymmetric and symmetric distributions.) The explanation was given as early as 1961 by Johansson (Ref. 3), who showed that the interaction of levels of opposite parity makes

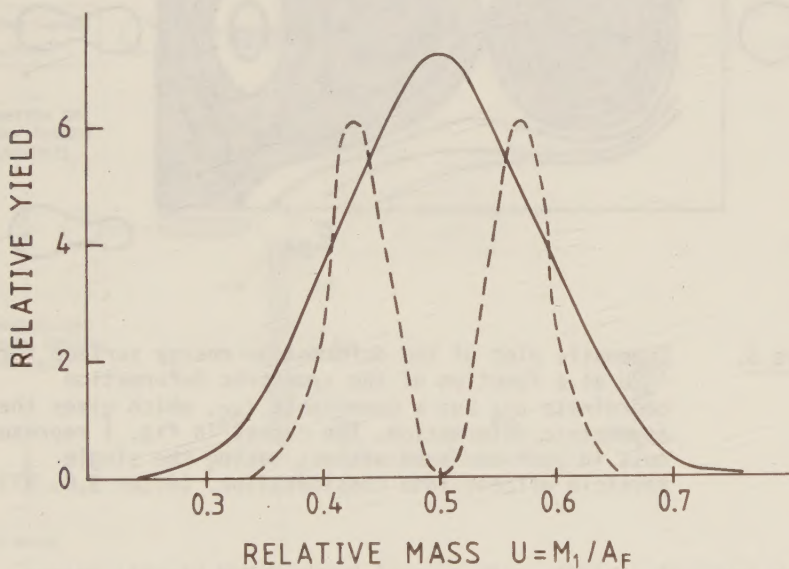


Figure 2. Examples of asymmetric mass distributions (^{252}Cf , spontaneous fission, dashed curve) and symmetric mass distributions (^{209}Bi , fission induced by 600 MeV protons, solid curve). A_F is the mass number of the fissioning nucleus.

the nuclei very soft to octupole (asymmetric) deformation. He found that for very elongated shapes, which are of interest in fission, stable octupole deformation occurs. The potential surface of the fission barrier will therefore possess two asymmetric valleys. Later more detailed calculations have confirmed this explanation. As an example the potential surface for ^{236}U is shown in fig. 3. Thus to explain the asymmetric mass distribution for heavy nuclei the single particle effects have to be taken into consideration.

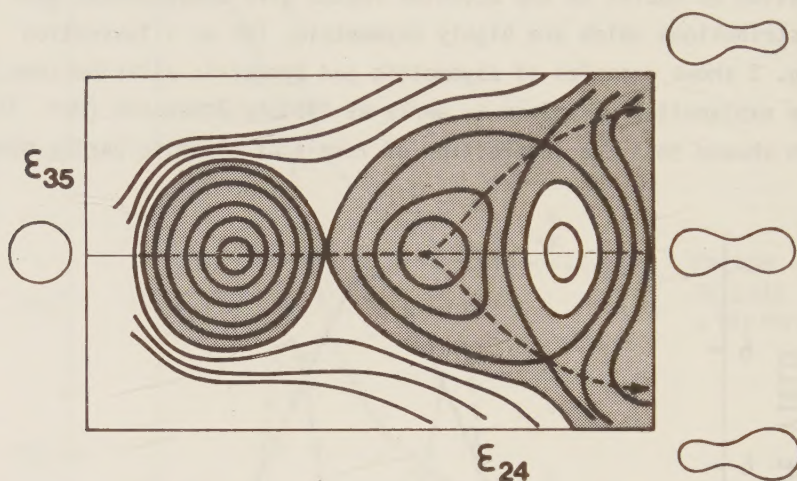


Figure 3. Schematic plot of the deformation energy surface for $^{236}_{92}\text{U}$ as a function of the symmetric deformation coordinate ϵ_{24} and a coordinate ϵ_{35} , which gives the asymmetric deformation. The curves in fig. 1 represent cuts in such surfaces without taking the single particle effects into consideration. (After S.G. Nilsson)

For an incident particle with several hundred MeV kinetic energy the process between the initial inelastic reaction and the fission products, which are experimentally detected, is rather complicated. Fig. 4 (from Ref. 4) illustrates the current picture of what happens.

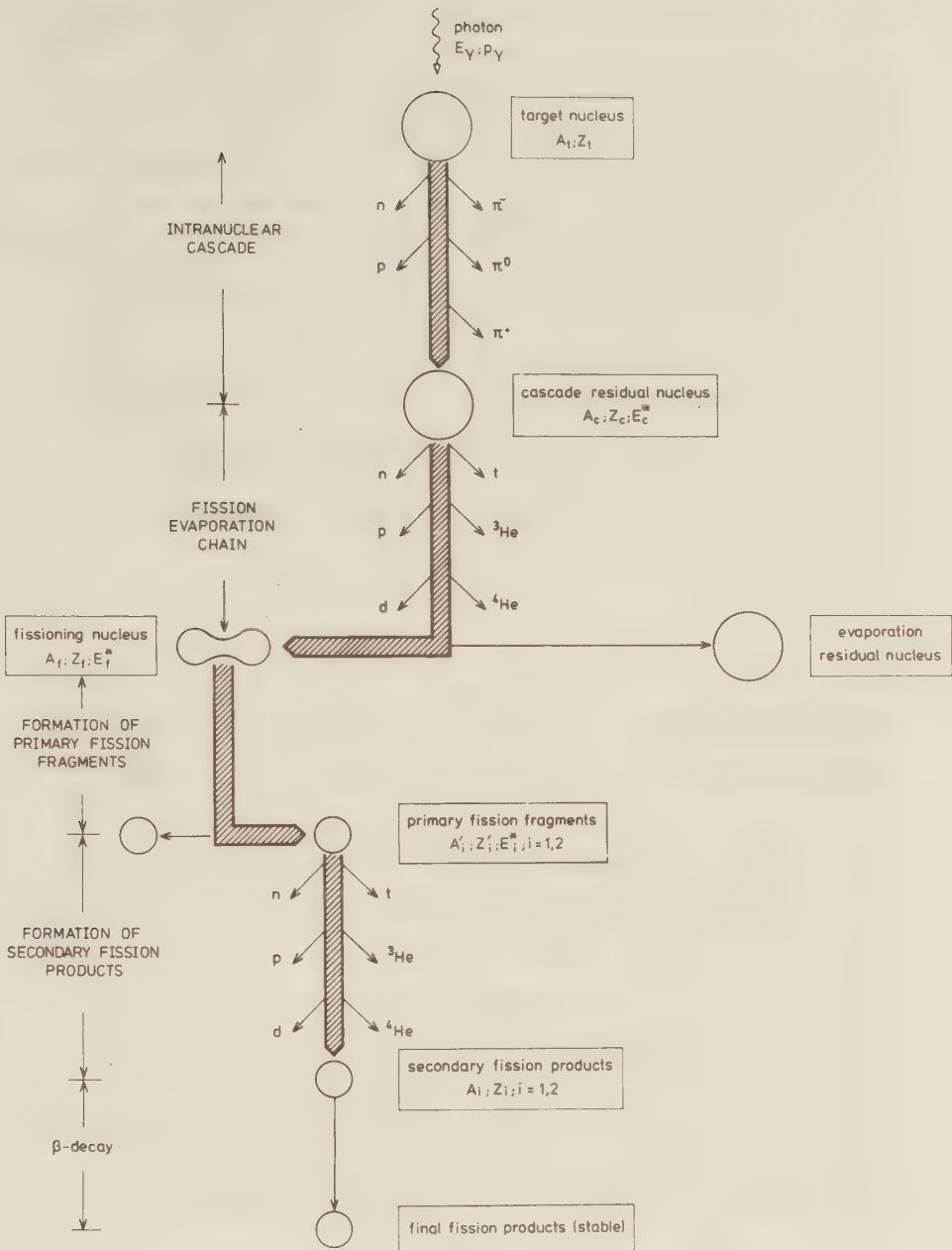


Figure 4. A model of what happens when a several hundred MeV particle hits a nucleus. (From Ref. 4.)

After the primary interaction between the incident particle and the target nucleus we have a fast intranuclear cascade ($\sim 10^{-22}$ sec). During this we have two-body collisions. Pions may be produced and interact with nucleons. Some particles may leave the nucleus. Afterwards we have what is called a cascade residual nucleus. This is characterized by mass number, charge and excitation energy. Fig. 5 shows the distribution of cascade residuals in the N-Z plane for 400 MeV photons and 600 MeV protons incident on $^{209}_{83}\text{Bi}$. (These energies are of interest in connection with this work.) We can see that we have a larger distribution for the protons. We further notice that the pion production is of greater importance for the photons. (For incident protons charge number 84 can be reached by adding the proton to the 83 protons of the Bi nucleus. But for incident photons a π^- must leave the nucleus to reach 84 protons.) Still mean values are very much the same

	A_{mean}	Z_{mean}	E^*_{mean}
600 MeV protons	205.8	82.7	158 MeV
400 MeV photons	205.9	82.3	162 MeV

If we compare the angular momentum of the cascade residuals we find for 600 MeV protons and Tb as target nucleus $7.5 \hbar$ (Paper V). The value for 400 MeV photons is approximately the same (Ref. 5).

After the cascade we have an energy equilibration. The nucleus deexcites by evaporating particles (n, p, d, t, ^3He , ^4He). Or it fissions. If so, the (primary) fragments are accelerated by the Coulomb interaction and they deexcite by particle evaporation (mainly neutrons) and γ -emission to give the fission products. Most of these are radioactive and decay by β^- and γ -emission until they reach the line of β -stability.

What information can we obtain experimentally that can be compared to theory? It depends on the experimental techniques used. With the double-kinetic-energy method we have used, we measure the

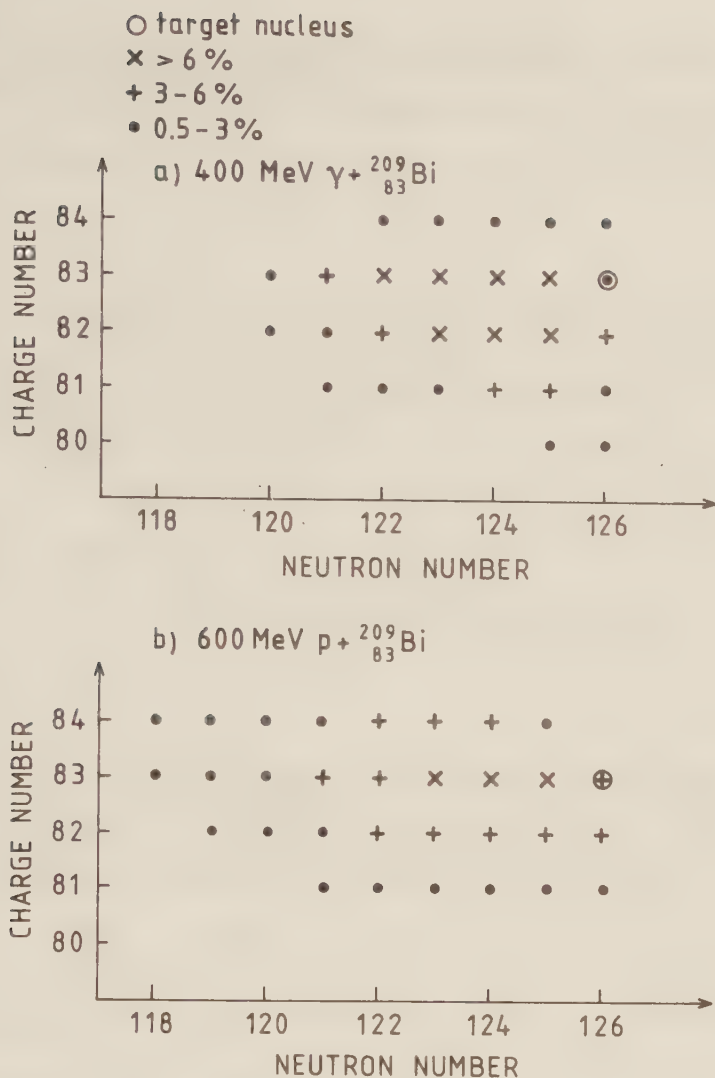


Figure 5. Distribution of cascade residuals in the N-Z plane for a) 400 MeV photons and b) 600 MeV protons incident on ${}^{209}\text{Bi}$. For each nuclide the symbol shows the relative number of cascade nuclei.

kinetic energy of the fission products. We obtain distributions for both the total kinetic energy and the single fragment kinetic energy. We obtain average single fragment kinetic energy and average total kinetic energy as a function of the mass and their standard deviations. We obtain mass distributions. From angular correlations between complementary fission products the momentum

and excitation energy distributions of cascade residuals leading to fission can be extracted. We determine cross sections. And we deduce fission barriers.

Thesis

The first part of the work was performed at the Lund electron synchrotron in 1974, using a bremsstrahlung beam of 600 MeV maximum energy. Thin targets (for details see ref. 6) were placed in a vacuum chamber and the fission products were registered in coincidence using Si surface barrier detectors. Later the vacuum chamber was moved to the 600 MeV proton synchrocyclotron at CERN and a series of experiments was performed in 1975 - 1979. Data were collected on line and stored event by event on magnetic tape for later analysis in Lund.

Paper I

In this experiment the double-kinetic-energy method was used for the first time in our laboratory. Apart from the results obtained we gained experience without which the experiments in papers III - VII could not have been performed. The experimental set up was tested with a ^{252}Cf source. This was also throughout all the experiments used to calibrate the detectors using both the fission fragments and the α -particles. The elements investigated were U, Th and Bi and as the rest of the targets used they were of the natural composition. The experiment was performed in the collimated bremsstrahlung beam of the Lund 1200 MeV electron synchrotron using 600 MeV electrons.

The bremsstrahlung beam contains photons of all energies up to the electron energy. We will thus have fission events induced by photons from the threshold up to the maximum energy. The distribution of events according to photon energy will depend on both the variation of the cross section with photon energy and the photon energy spectrum, which is roughly a $1/k$ spectrum, where k is the photon energy. For elements with low fission barriers, as U and Th, we

will thus have a large contribution from the giant resonance region, which is absent for Bi.

We determined the energy distributions both for the total kinetic energy and the single fragment kinetic energy. We further determined the averages of both the single fragment kinetic energy and the total fragment kinetic energy and their standard deviations as a function of the provisional mass. The mass distribution for Bi is compared to a liquid drop model calculation and activation analysis data. Good agreement is obtained. The results of this experiment show that for Bi we have symmetric fission.

Paper II

As mentioned in the introduction, if we have a particle with several hundred MeV kinetic energy incident on a nucleus, several things happen between the initial reaction and the detection of the fission products. These affect the angular correlation between complementary fission products. This correlation is also affected by geometric quantities. The calculations (computer code SOLID) reported in this paper give relations between angular correlations and momentum- and excitation energy distributions.

The free parameter in the calculations is the distribution of the component parallel to the beam direction of the linear momentum imparted in the cascade. This distribution is changed to obtain a good fit to the experimentally observed (in-plane) angular correlation. We then also get the ratio of the number of coincidences to the total number of fragments entering one of the detectors, which is used in the determination of the fission cross section. SOLID has proved very useful in experimental design too, as with an assumed momentum distribution we can see how the counting rate depends on the geometric configuration.

In an appendix some comments are given on the sensitivity of the calculated results to small changes in the values of different parameters used in the calculations, i.e. the cascade and evaporation momentum, the fissioning nucleus, the correlation between excitation energy and parallel and perpendicular momentum and the

mass loss during cascade and evaporation. The conclusion is that the calculated average values of the excitation energy and linear momentum imparted in the cascade are found insensitive to variations in the values of the various parameters used.

Paper III

It is of great interest to compare theoretical predictions and experimental results for the variation of both the fissility and the mass distribution with the fissioning nucleus. We then wanted to study elements lighter than Bi. The counting rate was however too low in the Lund bremsstrahlung beam so we continued the experiment with 600 MeV protons at CERN. The counting rate was now limited by the duty factor (0.5) and detector pile up. A disadvantage with protons compared to bremsstrahlung is that the interaction is not so wellknown. On the other hand we have monoenergetic particles incident on the target.

From now on we use a start (transmission) detector on one detector arm. In this and the next two papers it consisted of two $10 \mu\text{g}/\text{cm}^2$ thick carbon foils from which electrons were emitted on the passage of heavy reaction products. The electrons were partly collected on the entrance cone of a Channel Electron Multiplier tube. By measuring the flight time between this detector and the surface barrier detector behind it the mass of the fragment could be determined. In practice, however, under beam conditions the time resolution is so bad that with a flight path of 10 cm the mass determination is useless. But the time information from the start detector and the surface barrier detectors is quite crucial for sorting out the fission events from all other signals.

In this paper we report results on Tb and Ag. The fission cross sections and total kinetic energies were determined. The mass distributions are of symmetric shape.

Paper IV

We here report results from further runs on Ag and Tb and first runs on La. The symmetric mass distributions for Tb and Ag are confirmed. But for La we unexpectedly found indications of an asymmetric mass distribution. The number of events is small (125), so we decided to collect more data (see paper VI).

Fig. 6 shows the fission barrier heights for nuclei along the line of beta-stability calculated with three different macroscopic models. These are the liquid drop model (LDM, ref. 7), the modified liquid drop model (MLDM, ref. 8) and the droplet model (DM, ref. 9). The circles on the curves shown in fig. 6 indicate for each theory the approximate location of the so-called Businaro-Gallone point. Above this point the symmetric saddle is instable only towards fission, but below it becomes instable against two degrees of freedom, i.e. fission and mass-asymmetry. Below the Businaro-Gallone point the fragment mass-distribution is supposed to be highly asymmetric and above symmetric. The experimental mass distributions for Tb and La are in agreement with the predictions by theory. But for Ag we have disagreement as the increase in the width of the mass distribution is not seen experimentally.

Paper V

We have experimentally determined the coincidence rate as a function of the angle between the detectors for Ag, La and Tb. Using SOLID we can from this calculate the fissility as a function of excitation energy, which gives for each element a value of a_f/a_n , i.e. the ratio of the level density parameter at the saddle point and the ground state. In the fission-spallation competition calculations, we have two free parameters specifying the properties of the saddle point, i.e. the fission barrier height and a_f/a_n . With the latter determined we obtain the fission barriers, which turn out to be lower than predicted by macroscopic theories. It is interesting to note that the lower than normal value of the total fissility for La compared to Ag and Tb is not caused by a higher than normal

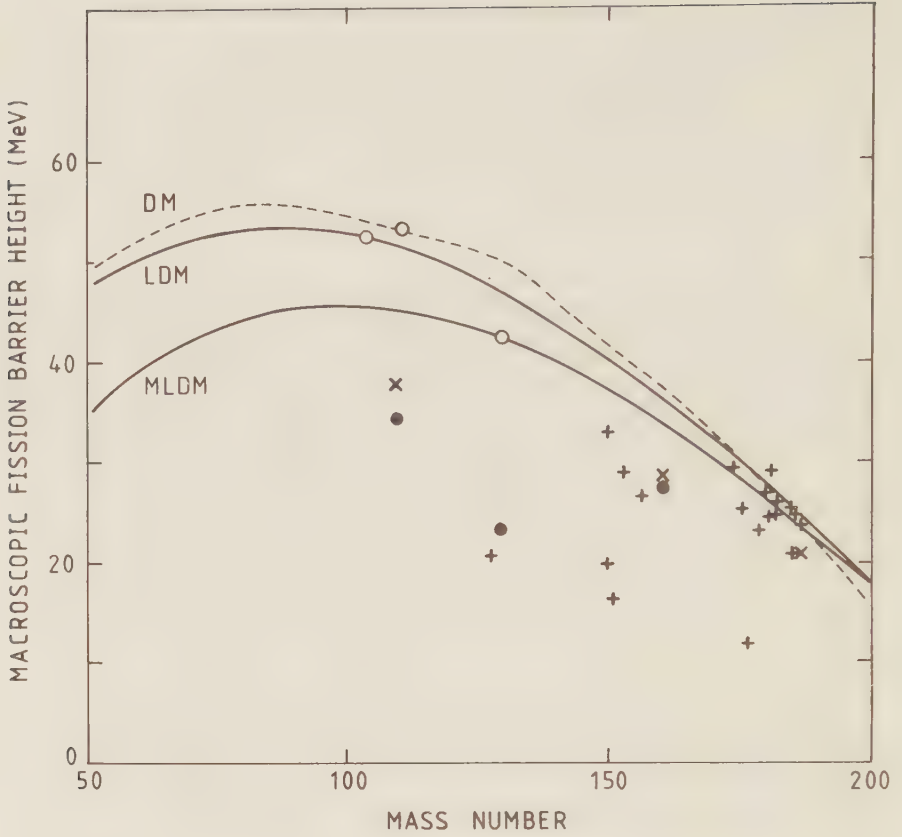


Figure 6.

The curves show the macroscopic fission barrier heights for nuclei along the line of beta-stability. The results of the three different theories are shown:

- LDM liquid drop model (ref. 7)
- MLDM modified liquid drop model (ref. 8)
- DM droplet model (ref. 9)

The circles on the curves indicate the positions of the Businaro-Gallone points in the theories. Also shown are experimental fission barriers from this work (●) and ref. 10 (+). Three barriers from the recent calculation of Möller and Nix (ref. 11) are shown as (x).

fission barrier but by a lower than normal value on a_f/a_n .

The total kinetic energies at symmetric mass divisions are higher than predicted by the liquid drop model and follow closely the Viola semi-empirical formula.

Paper VI

Two modifications of the experimental set up were made. A new start detector was used. It consisted of a $20 \mu\text{g}/\text{cm}^2$ carbon foil, placed at 45° with respect to the time-of-flight path, viewed by two channel electron multiplier plates. These are not so sensitive to radiation damage as the channel electron multiplier tube. The new start detector gave a larger solid angle too. Further a gasionization chamber was placed on the other detector arm. It consisted of 9.4 cm of 90% Ar - 10% CH_4 gas at a pressure of 20 torr for energy loss measurements. These as well as the measurements of the time difference between the anode of the gasionization chamber and the surface barrier detector behind it has been valuable for the sorting out of the fission events especially for the lightest target elements.

In this paper we give results for runs on Y, Sb and La. The total kinetic energies at symmetric mass division are still in good agreement with Viola's empirical relationship. The mass distributions for Y and Sb are symmetric, while the La mass distribution is broad with an indication of asymmetry. We then decided to run the neighbouring element Ce as well (paper VII). No indication is found of the Businaro-Gallone limit for target elements down to Se (unpublished data).

Paper VII

Our results on Ce and Yb are reported. For Yb we obtain a symmetric mass distribution. For Ce the mass distribution is broad, like the one we obtained for La, with a strong indication of asymmetry.

As discussed above fission of nuclei in the actinide region give asymmetric mass distributions. It is well-known that if we bombard

e.g. uranium with protons of increasing kinetic energy, the valley between the two peaks in the mass distribution is filled in (see fig. 7). We thus have a mixture of asymmetric and symmetric fission. The reason is that when we increase the proton energy, we increase the excitation energy of the fissioning nucleus. The asymmetric saddle remains. But the importance of the difference between the heights of the symmetric and the asymmetric barrier becomes less and the probability of symmetric fission increases.

The shape and width of the La and Ce mass distributions remind of the U mass distribution for 600 MeV protons. This indicates that the saddle is asymmetric. As there is a periodicity in the single particle levels the explanation could be the same as for the actinide region given above.

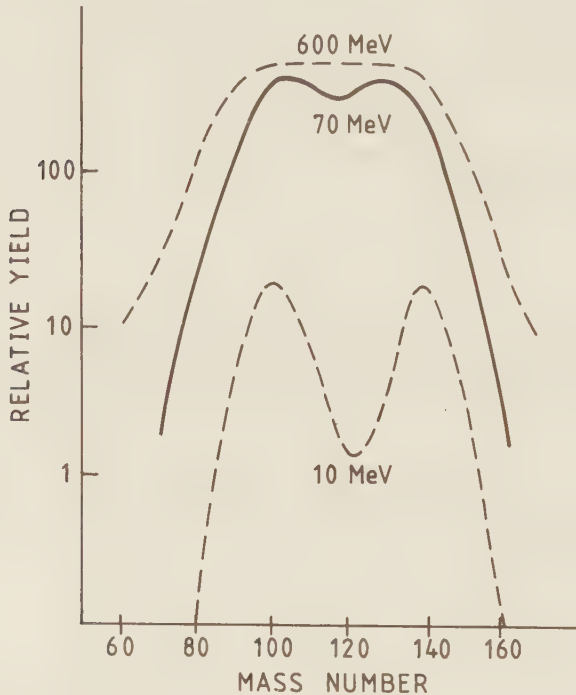


Figure 7. Schematic picture of the variation of the mass distribution with kinetic energy for protons incident on U.

Conclusions

When we compare the predictions of the macroscopic fission models with our experimental results we find several shortcomings of the models:

- 1) The predicted Businaro-Gallone point is not seen experimentally.
- 2) The predicted fission barriers are too high. In fig. 6 the filled circles are the fission barriers determined in this experiment. Some other experimental values taken from ref. 10 are also shown (+). In a recent calculation Möller and Nix (ref. 11) obtained fission-barrier heights of 28 nuclei. The three with the lowest mass numbers are shown in fig. 6 (x). The agreement with our experimental values for Ag and Tb is good.
- 3) The predicted total kinetic energies at symmetric mass divisions are too low. Our experimental values are in good agreement with the empirical relation of Viola.

We find broad mass distributions with an indication of asymmetry for La and Ce. We believe that this could be explained by single particle effects.

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With pleasure I take this opportunity to express my sincere gratitude to Professor Sven Johansson for introducing me into the field of nuclear science and for his support and encouragement during the course of this work.

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